The Detrital Origin of the Moncorvo Ordovician Ironstones

F. L. C. d'Orey

D.Sc., M.Sc., D.I.C., C.Eng., M.I.M.M.
Centro de Estudos Geológicos, Depart. 6 de Ciências da Terra, Faculdade de Ciências e Tecnologia, Quinta da Torre, 2825-114 Caparica, Portugal

ABSTRACT

Key words: magnetite; specularite; martite; detrital minerals; Moncorvo

The Moncorvo Ordovician ironstones in northeastern Portugal consist of iron ore sedimentary horizons frequently interbedded with psamites and quartzites. Ore reserves may probably exceed 1 000 million tonnes and this makes Moncorvo the largest iron ore deposit in the European Union. Compact poorly banded massive layers may exceed 90 meters in thickness which is quite an extraordinary feature for a Phanerozoic deposit. If the thickness of Precambrian deposits may reach a few hundred meters, the thickness of Phanerozoic deposits never exceed a maximum of 15 meters generally forming a number of comparatively thin layers confined to a particular member of a sedimentary sequence.

A detailed microscopic analysis of the ores revealed that initially a compact magnetite/quartzite layer, detrital in character (the magnetite occasionally showing chromite cores), was deposited by entrapment in near shore lagoons where rivers debouched, rather than in the open sea. This stage was followed by oscillating and transgressive shore lines which gave rise to breaks in sedimentation in combined river delta and shallow water marine environment where detrital material and fine iron oxide and clay suspensions were deposited in fluctuating environments. These events gave rise to layers of both magnetite (martite) and specularite intergrown with quartz, silicates and phosphates. Textural and mineralogical studies show that the deposits consist of ferruginous clastic sediments and are not chemically deposited cherts. Field, geological and palaeontological evidence also supports a detrital origin, the facies being typical of zones rich in oxygen and close to the feeding continent.

The uncommon huge development of Moncorvo was due to the fact that the deposits occur in restricted basins on a continental platform were clastic sediments were predominantly deposited. Not only morphologically but also chemically the deposits are more similar to Precambrian iron formations than to Phanerozoic ironstones.

RESUMO

Palavras chave: magnetite; martite; especularite; minerais detriticos; Moncorvo.

Os jazigos de ferro do Ordovícico de Moncorvo, situados no nordeste de Portugal, consistem de níveis sedimentares de minérios de ferro frequentemente interestratificados com psamitos e quartzitos. As reservas minerais provavelmente excedem 1 000 milhões de toneladas o que faz de Moncorvo o maior jazigo de ferro da União Europeia. Existem estratos compactos com mais de 90 metros de possaça o que é iníngulo em jazigos do Fanerozóico. Se a possaça de certos jazigos Precâmbicos de ferro pode atingir algumas centenas de metros a dos jazigos do Fanerozóico nunca excede um máximo de 15 metros sendo geralmente constituídos por um conjunto de estratos finos confinados a uma dada unidade de uma sequência sedimentar. O estudo microscópico pormenorizado dos minérios revelou que um estrato compacto caracterizado de detriticos constituído por magnetite e quartzo (em que a magnetite apresenta ocasionalmente núcleos de cromite) se depósitou em lagoas próximas da costa onde os cursos de água desaguavam e não num oceano. Seguidamente, linhas de praia oscilantes e transgressivas revelam interrupções de sedimentação em ambientes de zonas deltaicas e marinhas pouco profundas onde material detritico e suspensões de material ferruginoso e argiloso originaram estratos de magnetite (martite) e especularite interessados com quartzo, silicatos e fosfatos em ambientes fluctuantes. Os estudos texturais e mineralógicos demonstram que a origem dos jazigos é essencialmente clástica não se tratando de um chert ferrifero depositado quimicamente. Há também provas paleontológicas e de campo que apoiam uma origem detritica, cujas fácies são típicas de zonas ricas em oxigênio próximas dos continentes.

O enorme desenvolvimento de Moncorvo deve-se a que os jazigos ocorrem em bacias sedimentares restritas sobre uma plataforma continental onde sedimentos clásticos foram predominantemente depositados. Não só morfologicamente mas também quimicamente os jazigos de Moncorvo são mais semelhantes a jazigos Precâmbicos do que a jazigos Fanerozóicos.
INTRODUCTION

The Moncorvo Ordovician Ironstones are located east of the town of Moncorvo in northeastern Portugal forming a long ridge of iron bearing quartzites outcropping at the top of Reboredo Mountain Range and at Mua extending for 8 km and 0.850 km respectively (Figure 1). This ridge reaches an elevation of about 300 meters above the surrounding lowland which is 600 meters above sea level. At Mua the ore deposits form an assymetric syncline whereas at Reboredo Mountain Range they are part of the northern slope of the vast Moncorvo synclinorium (Figure 2). Numerous lens like ore bodies outcrop throughout the area but are not of sufficient size to be of economic importance.

The mines have operated for several periods since Roman times but the advent of the Bessemer techniques led to the abandonment of the mines due to the high phosphorus content of the ores. Sporadic works were conducted by two mining syndicates particularly during and after World War II but never on a large scale. Exploration and drilling carried out in recent years has shown the proven and probable ore reserves of Moncorvo to exceed 1,000 million tonnes with an average grade of 37 wt% Fe (Orey & Rebelo, 1983).

During the second half of this century the global distribution of iron ore products has changed dramatically, most of the traditional sources of Western Europe have been abandoned because of relatively low quality. During World War II, British iron ore containing between 18-25 wt.% Fe were used to sustain the war effort. Today’s economics demand blast furnace feed grades in excess of 60 wt.% Fe (Cohen, 1998).

Besides naturally rich ores (Australia, Brazil) some ores can be easily upgraded to meet the economic requirements and compete with the world market. For Europe using ores from Brazil half the ore price is attributable to transport charges (Cohen, 1998). Having this in mind, the exploitation of the enormous iron ore reserves of Moncorvo would be profitable if suitable concentrates could be produced.

Initial reports on feasibility studies have found the ores to be too enriched in deleterious materials (phosphorus) and abundance of silica. However, laboratory and semi-industrial testworks conducted by Cerveira (1978) and further mineralogical observations (Orey & Rebelo, 1983) proved that high intensity wet magnetic separation is viable in producing iron rich concentrates with phosphorus lower than 0.2 wt%. The microscopic investigations

Fig. 1. Geological map of the area around the Moncorvo ironstones (after Rebelo & Romano, simplified).
of the Moncorvo ironstones have shown that the mineralogy of iron and phosphorus minerals is extremely complex and that the different types of ores can not be classified by means of their physical appearance in hand specimens and/or by chemical assays (Orey, 1979, 1980; Orey & Rebelo, 1983). Because the ores are variable in mineralogy, grade and physical characteristics, routine microscopic examination of run of mine products and concentrates is essential in establishing the most suitable ore dressing flowsheet for the different sections of the deposits. Due to this complex intergrowth of iron and gangue minerals, lump products are not suitable for direct charging into the blast furnaces. The ores have to be finely ground and must be sintered prior to feeding.

In order to consolidate all the Moncorvo mining concessions the mines were nationalized in 1975. The aim of this operation was to develop an Iron and Steel Plan for Portugal and the exploitation of the Moncorvo iron ores was a priority in this scheme with a view to export to other European nations a substantial part of the concentrate production.

A slow progress of the Iron and Steel Development Plan in what concerns Moncorvo was ascribed to the lack of infrastructures for processing the ores as well as suitable and cheap transport for the concentrates. Both problems seemed to have been overcome when due to political reasons and external pressures (European Union) the Moncorvo Iron Ore Project was abruptly abandoned in 1988.

GEOLoGY

The area around Moncorvo consists of Lower Palaeozoic formations which were cratonized during the Hercynian Orogeny. It was during this orogeny that deformation and low grade regional metamorphism took place, followed by the intrusion of granites which preserved the pre-existent formations (Figure 1). After this orogeny a few episodes of more or less intense fracturing and block uplifting have taken place (Neiva, 1949, 1952; Thadeu, 1952; Duarte et al., 1966; Ribeiro & Rebelo, 1971; Rebelo & Ribeiro, 1977).

The "Douro Series" is a monotonous formation of shales and greywakes overlain by fossiliferous Ordovician rocks of detrital origin and Silurian sediments. Rebelo & Romano (1986) consider the Douro Series consisting of three units, the Pinhão, Rio Pinhão and Desejosa formations, to be wholly or partly Cambrian for a trilobite fauna was found in the uppermost unit of this series. The fossiliferous unit, the Desejosa formation, consists of slates and meta-greywacks overlaying the Rio Pinhão formation with meta-greywacks and meta-quartzogreywacks alternating with dark slates and the Pinhão formation with chloritic slates and thin metaquartzogreywacks. The Douro series are overlain by Ordovician detrital metasedimentary units such as Quinta da Ventosa formation essentially defined by its detrital character with quartz or quartz-pelitic clasts with a sericitic or pelitic matrix followed by the Quartzite formation which corresponds to the Armorican quartzites of Brittany and Iberia made up of well bedded
quartzites alternating with slates, the upper quartzite beds passing laterally into iron ores and finally the Slate formation consisting of a thick sequence of different types of slates in places containing pyrite cubes or pyritic nodules. The latter formation occupies the central part of the Moncorvo synclinorium. Two small Silurian outcrops consisting of fossiliferous bluish grey slates or graphitic slates which include lenticular beds of limestones were identified in the central part of the map.

The structure of the Moncorvo region is essentially due to the Hercynian Orogeny which gave rise to a series of complex folding striking E-W, the folds showing a series of axial depressions and culminations (Rebelo & Ribeiro, 1977). Thus, the Moncorvo synclinorium (Figures 1 & 2) shows a transversal depression which gave rise to the morphological appearance of an elongated basin trending E-W. In the central sector the axial inclinations are almost nil increasing gradually to the periclinal terminations where they reach 30°. The present structure of the Moncorvo synclinorium is due to the overlapping of two deformation phases although the first phase is responsible for the essential of that structure. Two types of left lateral shear faults trending NE-SW and NNE-SSW to N-S are frequently filled with quartz. The former shear faults are compatible with the field of tensions during the second phase of compression. The latter shear faults are parallel to the Great Vilarica fault to the west of Moncorvo that is known to be late Hercynian. Some of the shear faults are typically normal faults due to reactivation movements in a crustal tension system during the post Hercynian evolution of the Hesperic Massif (Ribeiro, 1974).

The area has been subjected to low grade regional metamorphism of the chlorite zone which has moderately affected the grainsize, mineralogy and banding of the ore deposits.

IRON ORE DEPOSITS

Four main iron ore deposits are considered: Pedrada, Reboredo and Carvalhosa in the Reboredo Mountain Range and Mua north of the latter (Figure 1).

As a whole the iron ore deposits consist of an Ordovician stratigraphic unit which thickness never exceeds 150 ± 20 meters and grades into quartzites along the strike. In the deposits of the Reboredo Mountain Range the iron ore horizons contain several bands of psamites and quartzites whereas at Mua there are no such bands the mineralized horizons consisting of an homogeneous poorly banded formation up to 90 meters thick. This is a most outstanding feature in a Phanerozoic ironstone for in other known deposits (Clinton, Wabana, Wisconsin, Cleveland, Northampton, Lorraine, Luxembourg, etc., Stanton, 1972) the mineralized beds never achieve thicknesses exceeding 15 meters and this seems to be the maximum for deposits showing true ironstone characteristics. Also, at the Reboredo Mountain Range, namely at Carvalhosa, 40 meters thick compact ore bands are quite common.

ORE PETROGRAPHY

An understanding of the nature of the iron bearing minerals and their association with gangue minerals is critical not only to the definition of processing options but also to the study of the genesis of the deposits. Microscopic, chemical and electronprobe analysis were used to classify the iron ores into broad genetic types.

Three main types of ores, classified in terms of mineralogy and texture, were identified:

1) Compact magnetite/quartzite ores

Irregular beds of magnetite/quartzite ores never exceeding 15 to 20 meters in thickness occur at the bottom of all main deposits (Figure 2). Similar types of ore outcrop at Sabor River 8 km NE of Mua, at Felgar 2 km north of Mua and at Felgueiras more than 2 km S of Reboredo. All these beds consist of equant interlocked grains of quartz and magnetite in a matrix of scarce chlorite, sericite and rare apatite. Occasional chromite cores were identified in magnetite (Figure 6a). Rare small rounded inclusions of common sulphides (pyrite, chalcopyrite, cubanite and pyrrhotite) as well as ilmenite were identified in magnetite. In these ores magnetite only shows incipient martitization even when outcropping (Figures 6b and 6c). It seems that an initially deposited bed of detrital quartz and magnetite sediments covered extensive basins with limits far exceeding the present limits of the Reboredo-Mua basins.
2) Granular marcite(magnetite)/specularite ores

Overlaining the previous formations, the marcite(magnetite)/specularite ores are the predominant source of iron ore. The ratios of marcite (with scarce relics of magnetite) to specularite are very irregular ranging from 14:86 to trace amounts of specularite the latter occurring only in the norther limb of the Mua syncline. Quartz, chlorite, sericite, chamosite, rockbridgeite, lazulite and other minor silicates and phosphates are present in the matrix. When specularite is predominant, (Figure 6d) the ores are characterized by a schistosity (SI) imparted by alternating thin laminations of platy specularite and quartz at high angle to the banding (SO) of marcite and coarse grained specularite A second phase of deformation along cleavage (S2) gave rise to the development of fine needle shaped specularite. Contrary to the compact magnetite/quartzite ores (whether out­cropping or not), magnetite in these granular marcite/specularite ores is mostly oxidized but martitization does not follow the usual triangular patterns being extremely irregular and patchy (Figure 6e). This peculiar type of oxidation is seen in samples from near surface down to more than 150 meters deep at Reboredo. It is obvious that this type of oxidation is not due to weathering.

3) Limonitic ores

Hydrous iron oxides of variable composition consisting of acicular goethite overgrown on rhythmic or botrioidal hematite are common features at the surface of the marcite(magnetite)/specularite ores (Figure 6f) Electronprobe micro­analysis has shown that this limonitic material in a detrital quartz matrix has adsorbed substantial amounts of phosphorus.

CHEMISTRY

The iron content of the Moncorvo ores is low, ranging in composition from 35 to 43 wt.% Fe with an average value of 37 wt.% Fe. Phosphorus is quite high ranging from 0.3 to 0.7 wt.%, some samples exceeding 1.2 wt.% P. Alumina is fairly constant with an average 6 to 8 wt.% Al2O3. The silica content is high, ranging from an average 27 wt.% SiO2 at Mua to an average 37 wt.% SiO2 in all Reboredo Mountain Range deposits. CaO, MnO, TiO2 and Cr2O3 are very low and total alkalis (K2O + Na2O) do not exceed 1.5 wt.%.

An Al-Si-Fe diagram (Figure 3) shows that the Moncorvo ironstones diverge chemically from other known Phanerozoic ironstones. Although more aluminous, the chemical composition of the Moncorvo ironstones makes them more similar to Precambrian banded iron formations than to Phanerozoic ironstones.

Low MgO and CaO (Figure 4) makes them again more similar to Precambrian banded iron formations.

GENESIS

The mineralogical system of Moncorvo consist of a complex intergrowth and distribution of mineral grains with important variations in their concentrations (Orey & Rebelo, 1983). Textural and mineralogical studies have shown that the most probable source material was a ferruginous clastic sediment and not a chemically deposited chert. Even in low metamorphic environment it is sometimes difficult to distinguish detrital quartz from chemically deposited chert (James, 1955). However, at Moncorvo, detrital grains of inclusion free quartz, outlined by a dust of iron oxides, were observed in a matrix of impure ferruginous quartz where specularite and iron bearing philosilicates were eventually developed under low metamorphic and diagenetic conditions (Figure 6g). Frequently, paralell strings of dust like particles of iron oxides were observed to traverse the whole area of the polished section; this may be regarded as indicating the existence of much larger grains of quartz and proves that a substantial part of the quartz is detrital in character. The existence of

<table>
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<th>Average Chemical Analysis of the Moncorvo Ironstones</th>
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<tr>
<td><strong>Reboredo</strong></td>
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<tr>
<td>SiO2</td>
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<tr>
<td>Al2O3</td>
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<tr>
<td>FeO</td>
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<td>P2O5</td>
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rounded magnetite grains with chromite cores of obvious initial magmatic formation also points to a detrital origin for this iron oxide. The “protolith” of these magnetite grains, some of them with chromite cores and inclusions of pyrrhotite, cubanite and chalcopyrite, is certainly magmatic in origin.

Consequently, the Moncorvo ironstones can by no means be defined as a dominant chemical or biochemical precipitate. It is more appropriate to classify them as a ferruginous quartzite defined by Dorr (1973) as a metasedimentary rock defined by Reboredo & Ribeiro (1977):

- transition of iron ores to quartzites along the strike
- occurrence of the same sedimentary structures in both iron ores and quartzites (cross bedding, load casts).
- presence of the same fossils in both iron ores and quartzites.

These facies are typical of the zones rich in oxygen and close to the feeding continent. Study of palaeocurrents and lateral variation of facies show transport from north to south. According to Ribeiro (1974), the continent which has fed with detritus the armorican sandstones would have been situated in the Middle Galicia - Eastern Trás-os-Montes zone of northwestern Iberia (Figure 5) and to the south of this area there was a shallow marine platform. It is considered that areas of relative tectonic stability with high peneplanation of the feeding continent are highly favourable to the formation of iron ore deposits. Thus, the geotectonic environment in which the Moncorvo deposits are localized and the nature of the associated rocks are not consistent with a deep marine chemical environment. The bottom layers of all economic deposits, as well as some uneconomic occurrences north and south of the Reboredo Mountain Range, consist of predominant clastic equant grains of magnetite and quartz. Separation of this clastic material could have taken place by its entrapment in lagoons where rivers debouched rather than in the open sea. Oscillating and transgressive shore lines gave rise to breaks in sedimentation in combined river delta and shallow water marine environments where detrital quartz and iron material as well as fine hydroxide suspensions and fine clay particles in different proportions were deposited. These correspond to the martite(magnetite)/specularite ores with different clay and iron silicates overlaying the quartz/magnetite deposits. The former are inter-
Rebelo & Ribeiro (1977), quote some authors who assumed a hypothetical oolith origin for the Moncorvo ironstones. Oolithic iron ores were initially identified by Neiva (1952) at Guadramil, some 90 km northeast of Moncorvo (Figure 5), the sideritic ooliths being associated with graphitic slates. This led Thadeu (1952) to conclude that the deposits at Guadramil were formed under reduced conditions whereas the Moncorvo ores that contain fossils of the shore line were deposited under oxidizing conditions. Oolithic textures were never found at the main Moncorvo deposits, (Mua, Carvalhosa, Reboredo and Pedrada). Ooliths are common in most iron ore deposits of the world including metamorphosed Precambrian banded iron formations. Oolithic textures are known to survive quite strong metamorphism which is not even the case of Moncorvo. James (1955) states that ooliths are preserved up to and including the sillimanite grade of metamorphism.

Other authors, still quoted by Rebelo & Ribeiro (1977) assume that regional metamorphism has transformed the original iron minerals into magnetite and late metamorphic events related to hydrothermal fluids associated to granite intrusion gave rise to martitization of magnetite and formation of specularite. This does not seem probable for the following reasons:

a) in the deepest zones of the deposits unaltered magnetite is the predominant iron oxide; if martitization was due to hydrothermalism the most affected zones would have been precisely the deepest zones.

b) introduced specularite of hydrothermal origin is not uncommon but it is small in quantity and generally easily distinguished on the basis of occurrence; at Moncorvo specularite is known to occur very far from the granite contact and is not restricted to the aureole of contact metamorphism.

The minerals in the martite (magnetite)/specularite zone were deposited in fluctuating environments with resultant interlayering and modification of contrasting materials and processes of both contemporaneous clastic sedimentation and closely following diagenesis. These have usually added some complications to the mineralogies that might have been expected from quiet and uninterrupted chemical precipitation.

Klein (1966), based on studies of several iron formations, states that the very frequent occurrences of quartz/magnetite/specularite rocks in which thin bands of magnetite alternate with thin bands of specularite (as in Figure 6h) indicate that the activity of the $O_2$ component was determined by the original assemblage and not by its metamorphic history; as such the $O_2$ component can not be considered as a perfectly mobile component. This is shown in the martite/specularite ores from near surface down to more than 150 meters deep where magnetite is very irregularly and patchy martitized (Figure 6e) due to varied oxidation ratios resulting from different oxygen contents of the rocks. These peculiar and atypical martitization textures are certainly not due to weathering. Also, the confinement of rocks of various oxidation ratios to well defined bands suggest that the differences in oxygen content are of premetamorphic origin. This conclusion is in harmony with the known fact that

b) Martitization of magnetite in outcropping magnetite/quartzite ores, showing the usual triangular pattern following (111) planes. Felgar.


d) Irregular and patchy martitization of magnetite in martite/specularite ores not showing the triangular oxidation network, Pedrada, D.D.H. S-4, 169 m.

Fig. 6

during regional metamorphism rocks in general behave as narrowly defined units closed to oxygen, the oxygen partial pressure in each unit being determined by the mineral assemblage and hence the original oxygen content rather than being externally imposed.

It was important for the uncommon huge development of the Moncorvo layers that these deposits occur in restricted basins on a continental platform were elasic sediments were predominanly deposited. Ordovician iron districts in northeastern Portugal represent different stages of facies gradation from oolithic iron ores of the Clinton type at Guadramil, as described by Neiva (1953) to a facies more similar to the Precambrian banded iron formations of the Superior type at Moncorvo.

The complete sequence at Moncorvo was as follows:

1) Deposition of detrital magnetite (with chromite cores), quartz, zircon and other detrital minerals in lagoons.
2) Deposition of magnetite, phosphates, clay and iron silicates in combined river delta and shallow water marine environment rich in oxygen.
3) Formation of coarse grained specularite and patchy oxidation (martitization) of magnetite at an initial diagenetic/early metamorphic stage.
4) First phase of deformation (D1) which gave rise to the genesis of acicular specularite.
5) Second phase of deformation (D2) with formation of further specularite, recrystallization of chertic quartz and formation of iron bearing silicates and phosphates; recrystallized quartz replaced iron oxides.

6) Post orogenic supergene alteration: the martite/specularite ores gave rise to goethite and the associated phosphorus minerals to green and brown rockbridgeite; the magnetite grains of the compact magnetite/quartzite ores when outcropping show the triangular oxidation pattern following (111) planes.

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